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High-concentration Er:YAG single-crystal fibers grown by laser-heated pedestal growth technique

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High-concentration Er:YAG single-crystal fibers have been grown using the laser-heated pedestal growth technique. Instability in the melt and concomitant opacity of fibers were observed at source concentrations higher than 15 mol.%. Spectroscopic examination shows that broadening of the linewidth of the $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition is strongly dependent on Er^{3+} concentration. © 2011 Optical Society of America

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Yttrium aluminum garnet (YAG) is used extensively as a host medium for rare-earth ion doped solid state lasers. Having a garnet structure, this crystal has many advantages as a host material for laser applications, including a high thermal conductivity to dissipate excess energy, the ability to incorporate a wide range of rare-earth ions, and the ease with which large, high-quality crystals can be grown. Burrus and Stone [1] were the first to demonstrate an Nd:YAG single-crystal fiber laser grown by the laser-heated pedestal growth (LHPG) technique. The fibers, pulled at 5 mm/min, were of a high optical quality and free from microcracks. Although Nd is the most common rare-earth ion used with YAG, other rare-earths such as Ho [2], Tm [2], Yb [3], and Er [4] have also been demonstrated to give laser action. In recent years, YAG single-crystal fibers have also been used in the medical field for laser thermotherapy [5]. A high chemical resistance, good mechanical strength, and high temperature tolerance makes them very attractive in the medical field.

The work presented in this Letter focuses on the production of YAG single-crystal fibers doped with high concentrations of Er^{3+} ions and the characterization of their spectroscopic properties. Because of the relatively low solubility of Er^{3+} ions in silica, conventional glass-based erbium-doped fiber amplifiers are doped at levels of approximately 1 or 2 mol.%—above this, the problems of clustering, nearest neighbor energy transfer, and concentration quenching become evident. As shown in this work, YAG is an excellent host material for this particular rare-earth ion at concentration levels as high as 10 mol.% when grown by LHPG.

Source rods for fiber production were made from a stoichiometric mixture of Y_2O_3 and Al_2O_3 powders. The powder mix was thoroughly agitated to ensure homogeneity before the addition of an appropriate concentration of Er_2O_3 powder and a final mixing stage. The resulting powder was then cold pressed at a force of 15 tons in a 16 mm diameter die. Pellets of approximately 1 mm thickness were then sintered at 1500 K for 12 h prior to being cut to produce square section source rods of dimension 0.9 mm × 0.9 mm × 15 mm. Sintering of the pellets was found to have only a marginal effect on improving sample density and great care was needed during the cutting process due to the fragility of the pellets.

An undoped, $\langle 111 \rangle$ -oriented crystalline sample of YAG was used as a seed crystal to initiate fiber growth and determine the crystallographic orientation of the final fibers. Fibers were grown at a pull rate of 1 mm/min to optimize crystal quality and prevent the formation of microvoids.

Absorption and fluorescence measurements were taken using a 0.5 m monochromator. The signal from an indium gallium arsenide (InGaAs) photodetector mounted at the exit slit was input to a computer-controlled lock-in amplifier to improve signal quality. A quartz halogen broadband source was used during absorption measurements and, for fluorescence emission spectra and lifetime measurements, excitation was achieved using a laser diode pump source at 975 nm. Estimates for the fluorescence lifetime were made by curve-fitting averaged sample data captured by digital oscilloscope.

The Er^{3+} :YAG crystals were approximately 400 μm in diameter (using a diameter reduction of approximately 2:1 during growth) with lengths ranging from 30 to 40 mm. At growth rates higher than 1 mm/min, periodic striations on the surface of the fibers were observed. Subsequent optical inspection also revealed the formation of microvoids throughout the fiber cross section, leading to sample opacity and a concomitant deterioration in optical quality. A common incident CO_2 laser power of approximately 17 W was maintained during growth for samples of this size. High-concentration Er:YAG fibres have previously been produced using the micropulling-down method (up to 50 mol.%) with very good optical quality [6]. In the current study, it was found that growth from source rods doped at or below 10 mol.% Er_2O_3 proceeded without difficulty and produced high quality, transparent crystals. However, growth from source material doped at 15 mol.% and above led to unacceptable levels of melt instability and oscillation. Small regions of scattered microvoids appeared in the fiber even at extremely slow growth rates, and eventually these microvoids filled the entire fiber volume directly above the melt. Continuing the growth, the occurrence of microvoids was observed to decrease and to disappear after some time with the melt stabilizing and the growing fiber becoming clear once more. This behavior of stability/instability repeated

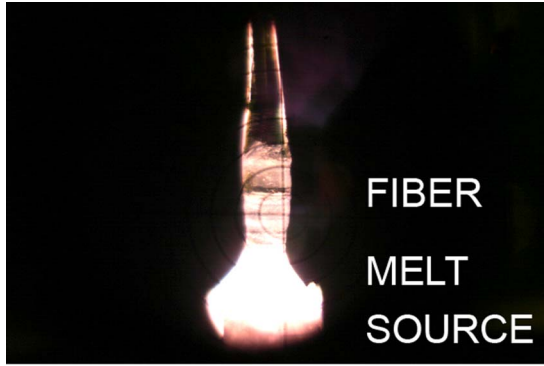


Fig. 1. (Color online) Er:YAG fiber grown from a source doped at 20 mol.% Er_2O_3 .

with a regular periodicity. Higher doping levels such as at 20 and 30 mol.% led to shorter periods between intervals of instability. As can be seen from Fig. 1, which shows a fiber being grown from a source rod containing 20 mol.% Er_2O_3 , the unstable melt causes a change of shape in the grown fiber and the fiber becomes almost completely opaque. It is anticipated that segregation at the freezing interface leads to an initial accumulation of Er at the interface and then, by thermocapillary convective mixing, throughout the melt volume. A substantial accumulation of dopant can be expected to influence not only the thermodynamic properties of the melt, but also the surface tension, which plays a key role in shape stability of the melt volume, and, in consequence, the relative rates of thermal absorption from the laser and radiative transfer away from the melt. Observations indicate that dopant accumulation proceeds to some critical value during instability and then freezes out, leading to a stable period of growth. In an effort to account for this, further studies will examine the axial distribution of Er^{3+} in the resulting crystal using electron microprobe analysis to determine segregation behavior during growth. This will help ascertain whether microcrystals of $\text{Er}_3\text{Al}_5\text{O}_{12}$ or ErAlO_3 have been precipitated, and if the crystal stoichiometry has been changed by such high doping levels, perhaps leading to the formation of a eutectic phase. Given the poor quality of samples grown from the most highly doped source material, subsequent spectroscopic analysis was performed only on fibers grown from source materials with concentrations of 0.5, 1, 5 and 10 mol.% of Er_2O_3 . By visual inspection, samples grown from 10 mol.% source material were seen to be pale pink in color.

Measurements of the absorption spectra of the Er^{3+} :YAG crystals were all conducted at room temperature along the fiber axes. Transmission measurements at a nonabsorption wavelength ($1.06\ \mu\text{m}$) indicated a consistent loss of approximately $0.2\ \text{cm}^{-1}$. Strong absorption in the range from 950 to 980 nm was observed for all fibers consistent with the $^4I_{15/2} \rightarrow ^4I_{11/2}$ transition of Er^{3+} . As can be seen from Fig. 2, which has been corrected for transmission loss, transitions to the $^4I_{13/2}$ manifold also produced broad absorption in the region from 1400 to 1700 nm. At dopant levels of 0.5 and 1 mol.%, absorption peaks are seen to be sharp and can be clearly distinguished. As may be anticipated, the integrated absorption intensity was observed to increase with increasing dopant concentration. The relative absorption between

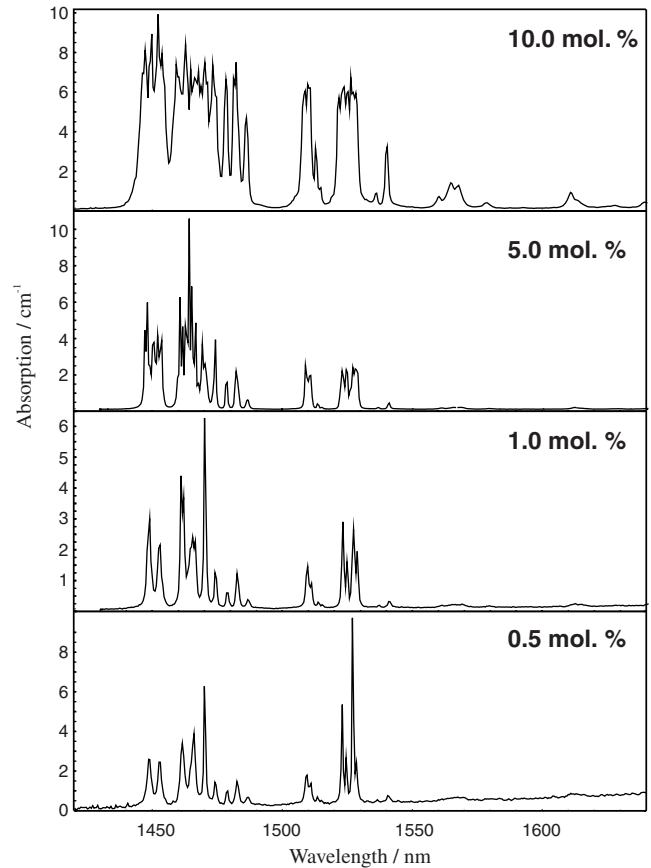


Fig. 2. Absorption spectrum of Er:YAG.

the bands centered around 1460 and 1530 nm is seen to change markedly with increased concentration of the Er^{3+} . Increasing dopant concentration is also seen to broaden absorption peaks, and at 10 mol.% (Fig. 2), the spectrum has been broadened to such an extent as to obscure its structure. Also of note in this case is the considerable increase in relative absorption centered at 1565 and 1612 nm to that shown in the other spectra. In comparison to previous work on Er:YAG crystal fibers grown by other methods, our results show a consistently lower absorption coefficient and, particularly at 10 mol.%, a markedly different spectral distribution [6]. This may well be due to the difference in growth methods with LHPG leading to a lower segregation coefficient and/or disruption to the YAG lattice structure at higher concentrations as suggested above.

In Er^{3+} :YAG, the main absorption transition from the $^4I_{15/2}$ energy level to the $^4I_{11/2}$ metastable level is used to populate the $^4I_{13/2}$ manifold. This provides a broad range of available excitation wavelengths with a peak around 970 nm. Fluorescence emission results from the radiative transition $^4I_{13/2} \rightarrow ^4I_{15/2}$. Figure 3 shows the axial fluorescence emission with varying levels of doping in the source material when samples were pumped using a diode laser operating at 975 nm. At lower concentrations (0.5 and 1 mol.%), many distinct emission peaks can be distinguished which result from Stark splitting of the $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition manifold. These spectra are consistent with previously reported work carried out on Er:YAG bulk crystals and single-crystal fibers [6–8]. The distinctive emission profile in the 1600 nm region

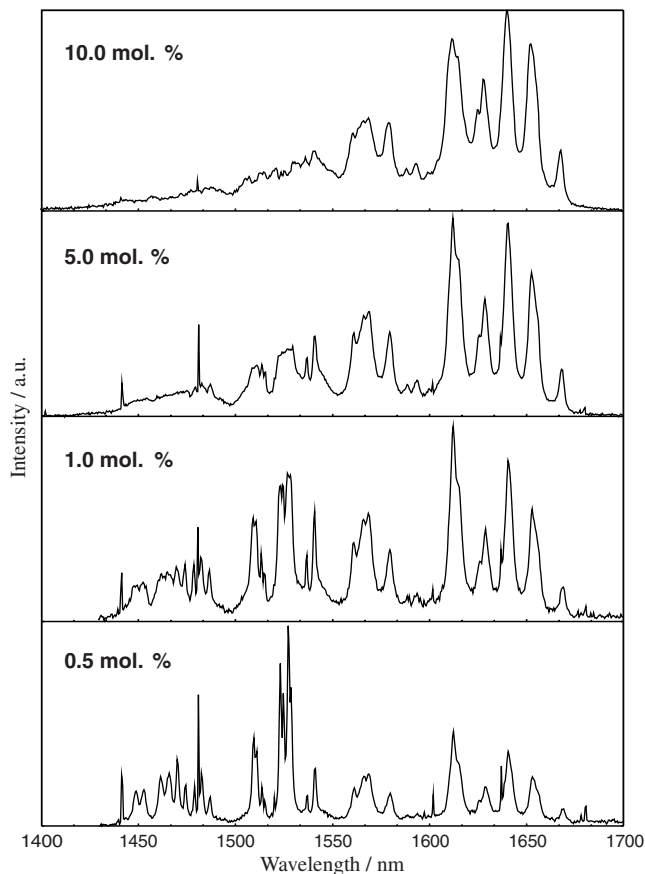


Fig. 3. Fluorescence spectrum of Er:YAG.

is also evident and is to be expected from high quality Er^{3+} doped YAG.

As the level of dopant concentration in the source material increases, we see a decrease in the relative emission intensity from the 1450–1550 nm region when compared to that from the 1600 nm region, in part due to self-absorption which we might expect from the results in Fig. 2. At 5 and 10 mol.% doping levels, the emission across the 1400 to 1600 nm region is considerably broadened with the peaks from individual Stark levels overlapping to form a broad emission bandwidth. Fluorescence at longer wavelengths retains distinguishable, relatively sharp Stark levels, even in samples grown from high-concentration source material. Broadening of the fluorescence spectrum is likely to arise, not only from increased nearest-neighbor effects, but also from increased disturbance to the lattice structure [9], particularly at very high doping levels.

As with the previous spectroscopic characterization, the integrated fluorescence lifetime measurements for the $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition were carried out at room temperature. A longpass filter was used to block wavelengths below 1250 nm. Lifetime measurements were taken both along and transverse to the fiber axis—no significant difference was found. Similarly, variation of

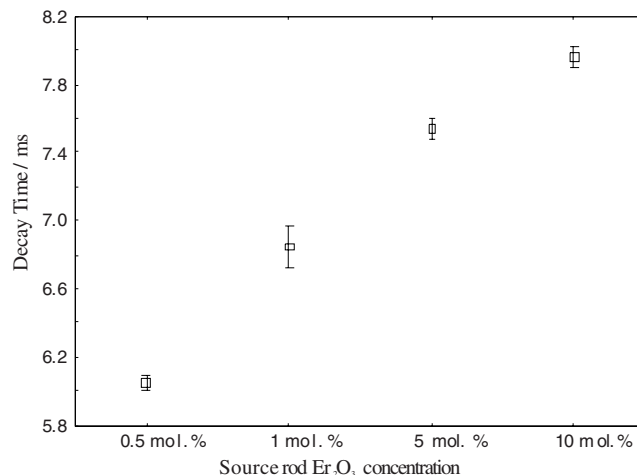


Fig. 4. Fluorescence decay lifetime of $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition.

pump power at 975 nm (from 20 to 100 mW) had no discernible effect on emission lifetime. As shown in Fig. 4, fluorescence lifetime increased with the increased dopant concentration. The lifetime of samples doped at 0.5 mol.% was found to be 6.05 ms and is in good agreement with previously reported work [6]. Samples doped at high concentrations (10 mol.%) exhibited lifetimes of 7.95 ms, again consistent with previous reports [10].

Er:YAG single-crystal fibers with a wide range of dopant concentrations were successfully grown using the LHPG technique. Spectroscopic analysis has shown that with increasing concentration, broadening of both absorption and fluorescence linewidths is observed. An increase in the fluorescence lifetime is also evident. At very high doping levels, crystal quality is considerably reduced and increased parasitic absorption across the emission spectrum is also evident.

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